





INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification ⁶:
C02F 1/461

A1

(11) International Publication Number: WO 98/58880

(43) International Publication Date: 30 December 1998 (30.12.98)

(21) International Application Number:

PCT/GB98/01864

(22) International Filing Date:

25 June 1998 (25.06.98)

(30) Priority Data:

97110693

25 June 1997 (25.06.97)

RU

(71) Applicant (for all designated States except US): STER-ILOX TECHNOLOGIES INTERNATIONAL LIMITED [GB/GB]; 85e Milton Park, Abingdon, Oxfordshire OX14 4RY (GB).

(72) Inventors; and

- (75) Inventors/Applicants (for US only): POPOV, Alexey Yurievich [RU/RU]; Solovini proezd, 2-353, Moscow, 117593 (RU). POPOV, Dmitriy Alekseyevich [RU/RU]; Solovini proezd, 2-353, Moscow, 117593 (RU).
- (74) Agents: VAUGHAN, Christopher, Tammo et al.; Haseltine Lake & Co., Imperial House, 15-19 Kingsway, London WC2B 6UD (GB).

(81) Designated States: AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GE, GH, GM, GW, HU, ID, IL, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW, ARIPO patent (GH, GM, KE, LS, MW, SD, SZ, UG, ZW), Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).

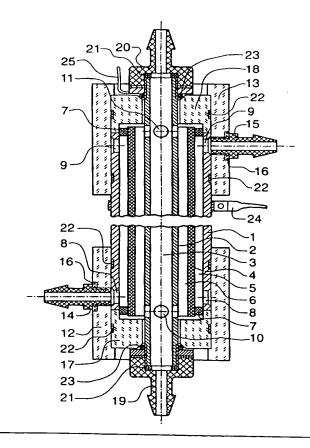
Published

With international search report.

(54) Title: METHOD AND APPARATUS FOR THE ELECTROCHEMICAL TREATMENT OF WATER AND AQUEOUS SALT SOLUTIONS

(57) Abstract

A method and apparatus for the electrolytic treatment of aqueous salt solutions, the apparatus comprising a diaphragm midstream electrolytic cell having coaxial tubular outer and inner electrodes (1, 2) separated by a semi-permeable diaphragm (4) so as to define an outer, working chamber (5) and an inner, auxiliary chamber (6). The inner electrode (2) is provided with apertures (10, 11) at either end allowing communication between the auxiliary chamber (6) and the interior (3) of the inner electrode (2). A working aqueous salt solution is passed upwards through the working chamber (5), while the auxiliary chamber (6) and the interior (3) of the inner electrode (2) is filled with an auxiliary aqueous salt solution. In use, a current is applied between the electrodes (1, 2), and gas bubbles formed in the auxiliary chamber (6) rise upwards and cause the auxiliary solution to circulate around the auxiliary chamber (6) and the interior (3) of the inner electrode (2) by way of the apertures (10, 11).



THIS PAGE BLANK (USPTO)

10

15

20

25

30

35

METHOD AND APPARATUS FOR THE ELECTROCHEMICAL TREATMENT OF WATER AND AQUEOUS SALT SOLUTIONS

The present invention relates to the electrochemical treatment of water and aqueous solutions of salt with the aim of altering the oxidising and reducing properties of the water or the aqueous solutions of salt. Such electrochemical treatment may take the form of anodic treatment for obtaining disinfectant solutions, cathodic water-softening treatment, or other treatments.

It is known from EP 0 394 232 to provide an electrolytic cell in which a tubular cathode is positioned within a tubular anode. An additional anode, in the form of a rod, is positioned within the tubular cathode. The anodes and the cathode are divided from each other by dielectric spiral elements. The apparatus is relatively compact, since the water is effectively subjected to electrolytic treatment in two electrolysis cells, one located inside the other.

It is known from GB 2 253 860, the disclosure of which is incorporated into the present application by reference, to treat water by passing this through an electrolytic cell having anode and cathode flow chambers separated by a semi-permeable membrane, one of the chambers being a working chamber through which water to be treated passes in an upward direction, and the other being an auxiliary chamber, which is in closed communication with a gas-separating chamber located at a higher level than the electrolytic cell. The electrolytic cell comprises a tubular outer electrode and a rod-shaped inner electrode, the two electrodes being concentric and separated by the semipermeable membrane so as to define the working and auxiliary chambers. Notwithstanding the semi-permeable membrane, the working and auxiliary chambers are

10

15

20

25

30

35

hermetically isolated from each other by way of elastic separator collars and each have entry apertures in the lower part and exit apertures in the higher part. Water having a higher mineral content than the water to be treated passes upwardly through the auxiliary chamber to the gas-separating chamber and recirculates to the auxiliary chamber by convection and by the shearing forces applied to the water through the rise of bubbles of gas which are generated on the electrode in the auxiliary chamber. In this way, the auxiliary solution circulates around a closed contour. The water pressure in the working chamber is higher than that in the auxiliary chamber, and gaseous electrolysis products are vented from the gas-separating chamber by way of a gas-relief valve. Some of the working solution will tend to pass from the working chamber to the auxiliary chamber across the semi-permeable membrane by virtue of the pressure gradient between the working and auxiliary chambers. Surplus auxiliary solution may be removed by way of the gas-relief valve or from the gas-separating chamber.

This method allows the pH value of the water being treated to be reduced from 7 to around 2 when the anode chamber is used as the working chamber. If instead the cathode chamber is used as the working chamber, the pH value of the water to be treated can be increased to around 12. This known method of electrolytic treatment is applied only to water having a relatively low concentration of dissolved salts and minerals (less than $10gdm^{-3}$), and the electricity supplied for the electrolytic treatment of water in the working chamber is only around 200 to 3000Cdm⁻³. Because the water to be treated has such a low concentration of dissolved salts and minerals, there is consequently a low concentration of useful electrolysis products (such as the chlorate (I) ion ClO which is produced when a

10

15

20

25

30

35

sodium chloride solution is used in the auxiliary chamber and which acts as a disinfecting agent). addition, water with a low concentration of salts and minerals tends to have a high ohmic resistance, which means that energy is used inefficiently when performing electrolysis. Furthermore, the small amount of electricity (200 to 3000Cdm⁻³) applied to the water in the working chamber is insufficient to ensure the full transformation of the ions of dissolved salts (such as chloride ions Cl) into useful electrolysis products (such as chlorate (I) ions ClO⁻). The incomplete electrolysis of dissolved salts means that a greater than theoretically necessary amount of salt must initially be dissolved in order to provide a required concentration of electrolysis products. This excess of dissolved salt can mean that the output of the electrolytic cell is overly corrosive, and when used as a disinfectant wash, tends to leave a coating of crystalline salt on surfaces which have been washed.

Furthermore, the prior art apparatus of GB 2253860 is of relatively cumbersome size and has a large number of connecting pipes and separate elements, which can result in low efficiency and reliability due to the likelihood of solution leakage.

It is useful to consider the basic chemical reactions which take place in the anode and cathode chambers of the electrolytic cell. If the working chamber contains the anode, then the following reactions take place:

Chloride ions transform into gaseous chlorine at the anode in accordance with the following equation:

2Cl.
$$\rightarrow$$
 Cl₂ + 2e⁻

Gaseous chlorine dissolves in water and forms hypochlorous acid in accordance with the following equation:

$$Cl_2 + H_2O \rightarrow H' + Cl' + HClO$$

10

15

20

25

30

35

Electrolysis of water also takes place in the anode chamber. The equation is as follows:

$$2H_2O \rightarrow 4H^+ + O_2 + 4e^-$$

As a result of this reaction, gaseous oxygen is liberated and the water becomes saturated with hydrogen ions. Consequently, the pH of the water falls in the anode chamber. The solubility of chlorine in the water reduces as the pH is lowered, and gaseous chlorine is liberated with oxygen.

Electrolysis of water takes place in the cathode chamber. The equation is as follows:

 $2H_2O + 2e^- \rightarrow 2OH^- + H_2$

Consequently gaseous hydrogen is liberated at the cathode, and the concentration of hydroxide ions rises, thereby increasing the water pH in the cathode chamber.

It follows from this analysis that the oxidizing ability of water is determined by the concentration of hypochlorous acid, and the reduction ability by the concentration of hydroxide ions. Water which has been under electrolytic treatment according to the method described in GB 2253860 has a low concentration of hypochlorous acid and hydroxide ions due to the low mineralisation of the initial water.

One way of estimating the effectiveness of a sterilising solution produced by the electrolytic treatment of a salt solution is to measure the concentration of "free chlorine", by which is understood the concentration of hypochlorous acid in water and the concentration of the chlorate icn (formed by the dissociation of hypochlorous acid).

The concentration of free chlorine in water which has been treated in the anode chamber of the electrolytic cell of GB 2253860 does not usually exceed 0.2 to 0.6gdm⁻³, although the solubility of gaseous chlorine in water is much higher (7.3gdm⁻³ at 20°C). It is therefore apparent that water which has been under

10

15

20

25

30

35

electrolytic treatment in accordance with the known method has a concentration of free chlorine not more than 3 to 10% of the possible maximum. This low concentration is a result of the fact that only a small proportion of the chloride ions which are drawn across the permeable membrane from the cathode flow chamber to the anode flow chamber actually reach the anode to be combined so as to form gaseous chlorine. Most of the chloride ions in the anode chamber are carried out of the electrolytic cell with the output of the anode flow chamber before reaching the anode itself.

According to a first aspect of the present invention, there is provided an apparatus for the electrolytic treatment of aqueous salt solutions, the apparatus comprising an electrolytic cell having a first, outer, generally tubular electrode and a second, inner, generally tubular electrode, the first and second electrodes being disposed substantially coaxially and separated from each other by a generally tubular semi-permeable membrane also disposed substantially coaxially with the electrodes so as to form an outer, working chamber defined as the volume bounded by the inner surface of the outer electrode and the outer surface of the semi-permeable membrane and an inner, auxiliary chamber defined as the volume bounded by the inner surface of the semi-permeable membrane and the outer surface of the inner electrode, the electrodes having upper and lower ends when the electrolytic cell is positioned vertically, characterised in that the tubular wall of the inner electrode is provided with exit apertures at its upper end region and entry apertures at its lower end region end region allowing communication between the inner, auxiliary chamber and the interior of the inner electrode.

According to a second aspect of the present

10

15

20

25

30

35

invention, there is provided an apparatus for the electrolytic treatment of aqueous salt solutions, the apparatus comprising a generally vertical midstream diaphragm electrolyser containing an outer tubular electrode inside which an inner electrode is coaxially positioned, a tubular semi-permeable ceramic diaphragm coaxially positioned between the electrodes and dividing the inter-electrodal area into an inner and an outer electrode chamber which are hermetically isolated one from the other by dielectric separators, the outer electrode chamber being a working chamber connected to lines for inputting and outputting the aqueous salt solutions being treated, and the inner electrode chamber being an auxiliary chamber provided with a circulation contour and connected to lines for inputting and outputting an auxiliary solution, characterised in that the inner electrode takes the form of a hollow cylinder which in its lower part is connected through its hollow section with the auxiliary solution input line and in its upper part is connected through its hollow section with the auxiliary solution output line, and in that the inner electrode is provided with input and output apertures which connect the hollow section of the inner electrode with the auxiliary chamber and which form a circulation contour for the auxiliary solution.

According to a third aspect of the present invention, there is provided a method of treating aqueous salt solutions in a generally vertical electrolytic cell, the cell comprising a first, outer, generally tubular electrode and a second, inner, generally tubular electrode, the first and second electrodes being disposed substantially coaxially and separated from each other by a generally tubular semipermeable membrane also disposed substantially coaxially with the electrodes so as to form an outer,

10

15

20

25

30

working chamber defined as the volume bounded by the inner surface of the outer electrode and the outer surface of the semi-permeable membrane and an inner, auxiliary chamber defined as the volume bounded by the inner surface of the semi-permeable membrane and the outer surface of the inner electrode, the inner electrode being provided with apertures at its lower and upper ends allowing communication between the inner, auxiliary chamber and the interior of the inner electrode, wherein:

- i) a working aqueous salt solution is supplied to a lower end of the working chamber by an input line and taken from a higher end of the working chamber by an output line;
- ii) an auxiliary aqueous salt solution is supplied to a lower end of the inner electrode by an input line so as substantially to fill the interior of the inner electrode and the auxiliary chamber by way of the apertures;
 - iii) an electric current is applied between the outer and the inner electrodes;
 - iv) gases formed by electrolysis in the auxiliary chamber are allowed to rise and escape through the apertures provided at an upper end of the inner electrode and are passed out by way of an output line provided at an upper end of the inner electrode; and
 - v) auxiliary solution in the auxiliary chamber is driven by bubbles of said gases and passes upwards through the auxiliary chamber, through the apertures at the upper end of the inner electrode, down through the interior of the inner electrode, and back into the lower end of the auxiliary chamber through the apertures at the lower end of the inner electrode.

Because the combined volume of auxiliary solution
and gases which is output per unit time from the
auxiliary chamber is generally greater than the volume

10

15

20

25

30

35

of auxiliary solution which is input per unit time, the total area of the exit apertures in the inner electrode is advantageously greater than that of the entry apertures.

It is also advantageous for the exit apertures to be configured so as to impart a tangential velocity component to the auxiliary gas-liquid mixture as this passes from the upper end of the auxiliary chamber into the hollow section of the inner electrode. This may be done by forming the apertures such that their axes are not disposed radially with respect to the inner electrode, but are angled relative thereto. In this way, a cyclonic flow into the hollow section of the inner electrode is achieved, with the heavier liquid component of the auxiliary gas-liquid mixture being urged towards the wall of the inner electrode by centrifugal force, and the lighter gas component conversely being urged towards the central axis of the inner electrode, thereby assisting the gas-liquid separation process.

The semi-permeable diaphragm may be made of a ceramic material, such as zirconium oxide.

Embodiments of the present invention are more compact than the prior art apparatus and have fewer and shorter fluid connecting lines, thereby reducing leaks and pressure losses and thereby providing enhanced reliability and efficiency. This is a result of the auxiliary solution circulation contour being located internally, i.e. the external expansion tank of the prior art is replaced by the interior of the hollow inner electrode, with the connecting pipes in the auxiliary solution circulation contour being replaced by the apertures in the wall of the inner electrode.

According to a fourth aspect of the present invention, there is provided a product solution produced by the method of the third aspect as defined

10

15

20

25

30

35

hereinabove.

For a better understanding of the present invention, and to show how it may be carried into effect, reference shall now be made by way of example to the accompanying drawing, in which:

FIGURE 1 shows a vertical cross-section through an embodiment of the present invention.

The electrolytic cell of Figure 1 includes an outer tubular electrode 1, a coaxially positioned inner electrode 2 having a hollow section 3, and a semi-permeable ceramic diaphragm 4 coaxially positioned between the electrodes 1 and 2. A working chamber 5 is defined as the space bounded by the inside of the outer electrode 1 and the outside of the ceramic diaphragm 4, and an auxiliary chamber 6 is defined as the space bounded by the inside of the ceramic diaphragm 4 and the outside of the inner electrode 2. The working and auxiliary chambers 5,6 are hermetically isolated one from the other (notwithstanding the semi-permeable ceramic diaphragm 4) by elastic, dielectric separators Entry apertures 8 are provided in the wall of the outer electrode 1 for supplying aqueous salt solution to the working chamber 5 (the entry apertures 8 being positioned in the lower part of the outer electrode 1). Exit apertures 9 are provided in the upper part of the outer electrode 1 and are used for letting processed aqueous salt solution out of the working chamber 5.

The inner electrode 2 is also provided with entry apertures 10 (in the lower part) and exit apertures 11 (in the upper part). The former are used for supplying auxiliary solution into the auxiliary chamber 6, and the latter for allowing processed auxiliary solution to be removed from the auxiliary chamber 6. The outer electrode 1 is disposed between lower and upper dielectric blocks 12,13. A connection 14 is positioned in the lower block 12 for supplying aqueous salt

solution to the working chamber 5 via the entry aperture 8. A feed pipe (not shown) is joined to the connecting pipe 14 and is used for bringing aqueous salt solution to the electrolyser. A connecting pipe .15 is positioned in the upper dielectric block 13 for 5 allowing processed aqueous salt solution to exit the working chamber 5 through the exit apertures 9. outlet pipe (not shown in the diagram) is joined to the connecting pipe 15 and is used for carrying the processed aqueous salt solution out of the 10 electrolyser. The connecting pipes 14 and 15 are hermetically sealed by elastic separators 16. inner electrode 2 is positioned within dielectric sleeves 17 (lower) and 18 (upper). Connecting pipes 19 15 (entry) and 20 (exit), positioned on the ends of the inner electrode 2, are hermetically sealed by elastic separators 21. The connecting pipe 19 is used for supplying the auxiliary solution into the hollow section 3 of the inner electrode 2. A feed pipe for the auxiliary solution (not shown) is joined to the 20 connecting pipe 19 and is used for supplying the auxiliary solution to the electrolyser. An outlet pipe for the auxiliary solution (not shown) is joined to the connecting pipe 20 and is used for letting the 25 processed auxiliary solution and electrolytic gases out of the electrolyser. In order to seal the working chamber 5 and the auxiliary chamber 6, the apparatus is equipped with elastic separators 22 and 23. electrolyser is provided with electrical terminals 24 30 and 25, which are positioned on the outer electrode 1 and the inner electrode 2 respectively, so as to permit connection of a current supply (not shown).

When the apparatus is in use, aqueous salt solution to be processed is supplied through the entry connecting pipe 14 and the entry aperture 8 so as substantially to fill working chamber 5. Auxiliary

35

aqueous salt solution is supplied through the entry connecting pipe 19 and into the hollow section 3 of the inner electrode 2. The auxiliary solution passes into and substantially fills the auxiliary chamber 6 by way 5 of the entry apertures 10. The supply of auxiliary solution is then cut off or reduced. The pressure of the working solution flowing through the working chamber 5 is set higher than the pressure of the auxiliary solution in the auxiliary chamber 6. 10 potential difference is applied across the terminals 24 and 25 so as to cause an electric current to flow between the outer and the inner electrodes 1,2 through the working and auxiliary solutions. This brings about electrolytic treatment of the working solution in the 15 working chamber 5, from which the treated solution leaves the electrolyser by way of the exit aperture 9 and the exit connecting pipe 15. The auxiliary solution is drawn by rising bubbles of electrolytic gases (the gas-lift) generated in the auxiliary chamber 20 6 during electrolysis into the upper part of the auxiliary chamber 2 and, together with the electrolytic gases, leaves the auxiliary chamber 6 through the exit apertures 11 and enters the hollow section 3 of the inner electrode 2. An expansion of the flow occurs in 25 the hollow section 3, whereby the electrolytic gases are separated from the auxiliary solution and leave the electrolyser through the exit connecting pipe 20. separated auxiliary solution is carried down to the entry apertures 10 and, under the action of the 30 discharge created by the gas-lift described above, is drawn through the entry apertures 10 into the auxiliary chamber 6. In this way, circulation of the auxiliary solution around a closed contour formed by the auxiliary chamber 6, the exit apertures 11, the hollow 35 section 3 of the inner electrode 2 and the entry apertures 10 is achieved. The volume of auxiliary

10

15

20

25

30

35

solution in the electrolyser is replenished, over time, by way of solution which filters through the semipermeable ceramic diaphragm 3 down the pressure gradient between the working and auxiliary chambers Any excess of auxiliary solution is taken from the electrolyser through the exit connecting pipe 20 together with the electrolytic gases. If it becomes necessary to provide additional auxiliary solution or to replace the auxiliary solution, this may be done by way of the entry connecting pipe 19. As a result of the formation of electrolytic gases, the volume of the gas and liquid mixture leaving the auxiliary chamber 6 is greater than the volume of the auxiliary solution entering the auxiliary chamber 6, and the overall area of the exit apertures 11 may be greater than the overall area of the entry apertures 10.

In order to obtain a more effective isolation of the electrolytic gases from the auxiliary solution in the hollow section 3 of the inner electrode 2, the exit apertures 11 may be disposed with a tangential as well as a radial component, i.e. the exit apertures 11 may be angled so as to impart a tangential velocity component to the auxiliary gas-liquid mixture as this passes from the auxiliary chamber 6 into the hollow section 3 of the inner electrode 2. The cyclonic motion thus imparted to the auxiliary gas-liquid mixture in the hollow section 3 of the inner electrode 2 tends to throw the heavier auxiliary liquid against the wall of the inner electrode 2 and concentrates the lighter gas components near the axis for removal from the electrolyser through the exit connecting pipe 20.

For most applications, the electrolytic cell is configured such that the outer electrode is an anode and the inner electrode a cathode. In this way, aqueous salt solutions can be electrolytically treated to obtain, for example, a disinfectant solution, with a

10

saturated solution of sodium chloride being used as the auxiliary solution. In some applications, the working solution has a higher mineral content, i.e. concentration of dissolved salts, than the auxiliary solution.

The apparatus may be arranged in alternative configurations, e.g. with the outer electrode 1 being the cathodé and the inner electrode 2 the anode, thereby enabling cathodic water softening or other processes to take place.

10

15

20

CLAIMS:

- An apparatus for the electrolytic treatment of aqueous salt solutions, the apparatus comprising an electrolytic cell having a first, outer, generally tubular electrode and a second, inner, generally tubular electrode, the first and second electrodes being disposed substantially coaxially and separated from each other by a generally tubular semi-permeable membrane also disposed substantially coaxially with the electrodes so as to form an outer, working chamber defined as the volume bounded by the inner surface of the outer electrode and the outer surface of the semipermeable membrane and an inner, auxiliary chamber defined as the volume bounded by the inner surface of the semi-permeable membrane and the outer surface of the inner electrode, the electrodes having upper and lower ends when the electrolytic cell is positioned vertically, characterised in that the tubular wall of the inner electrode is provided with exit apertures at its upper end region and entry apertures at its lower end region end region allowing communication between the inner, auxiliary chamber and the interior of the inner electrode.
- 2. An apparatus for the electrolytic treatment of aqueous salt solutions, the apparatus comprising a generally vertical midstream diaphragm electrolyser containing an outer tubular electrode inside which an inner electrode is coaxially positioned, a tubular semi-permeable ceramic diaphragm coaxially positioned between the electrodes and dividing the inter-electrodal area into an inner and an outer electrode chamber which are hermetically isolated one from the other by dielectric separators, the outer electrode chamber being a working chamber connected to lines for inputting and outputting the aqueous salt

solutions being treated, and the inner electrode chamber being an auxiliary chamber provided with a circulation contour and connected to lines for inputting and outputting an auxiliary solution, characterised in that the inner electrode takes the 5 form of a hollow cylinder which in its lower part is connected through its hollow section with the auxiliary solution input line and in its upper part is connected through its hollow section with the auxiliary solution 10 output line, and in that the inner electrode is provided with input and output apertures which connect the hollow section of the inner electrode with the auxiliary chamber and which form a circulation contour for the auxiliary solution.

- 3. An apparatus as claimed in claim 1 or 2, wherein the overall area of the exit apertures in the inner electrode is greater than the overall area of the entry apertures.
- 4. An apparatus as claimed in claim 1, 2 or 3, wherein the exit apertures in the inner electrode have major axes which are directed with a tangential component relative to radii of the generally tubular inner electrode.
 - 5. An apparatus as claimed in any preceding claim, wherein the outer electrode is an anode and the inner electrode is a cathode.
 - 6. An apparatus as claimed in any of claims 1 to 4, wherein the outer electrode is a cathode and the inner electrode is an anode.
- 7. A method of treating aqueous salt solutions in a generally vertical electrolytic cell, the cell comprising a first, outer, generally tubular electrode and a second, inner, generally tubular electrode, the first and second electrodes being disposed substantially coaxially and separated from each other by a generally tubular semi-permeable membrane also

OCID <WO 9858880A1 1 >

15

25

10

15

20

25

30

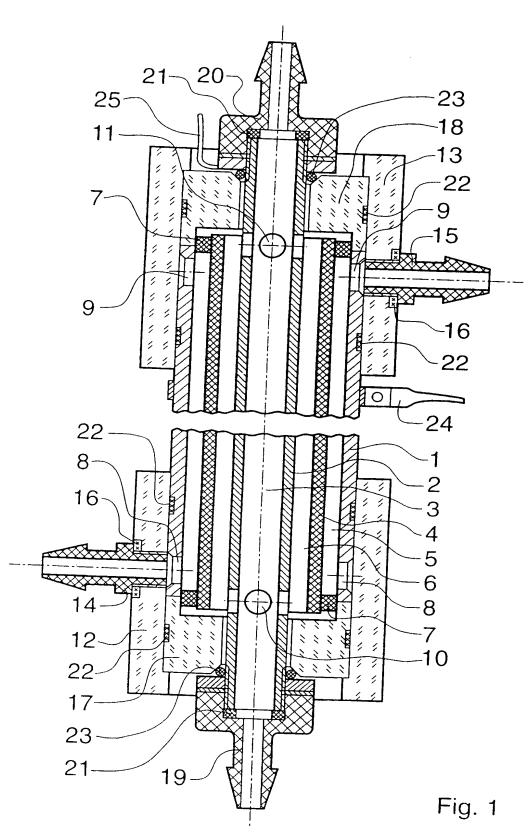
disposed substantially coaxially with the electrodes so as to form an outer, working chamber defined as the volume bounded by the inner surface of the outer electrode and the outer surface of the semi-permeable membrane and an inner, auxiliary chamber defined as the volume bounded by the inner surface of the semi-permeable membrane and the outer surface of the inner electrode, the inner electrode being provided with apertures at its lower and upper ends allowing communication between the inner, auxiliary chamber and the interior of the inner electrode, wherein:

- i) a working aqueous salt solution is supplied to a lower end of the working chamber by an input line and taken from a higher end of the working chamber by an output line;
- ii) an auxiliary aqueous salt solution is supplied to a lower end of the inner electrode by an input line so as substantially to fill the interior of the inner electrode and the auxiliary chamber by way of the apertures;
- iii) an electric current is applied between the outer and the inner electrodes;
- iv) gases formed by electrolysis in the auxiliary chamber are allowed to rise and escape through the apertures provided at an upper end of the inner electrode and are passed out by way of an output line provided at an upper end of the inner electrode; and
- v) auxiliary solution in the auxiliary chamber is driven by bubbles of said gases and passes upwards through the auxiliary chamber, through the apertures at the upper end of the inner electrode, down through the interior of the inner electrode, and back into the lower end of the auxiliary chamber through the apertures at the lower end of the inner electrode.
- 8. A method according to claim 7, wherein the auxiliary solution and said gases are given a cyclonic

motion upon entry into the interior of the inner electrode by means of the apertures at the upper end of the inner electrode being disposed with major axes having a tangential component relative to radii of the inner electrode.

 9 . A product solution produced by the method of claims 7 or 8.





SUBSTITUTE SHEET (RULE 26)

INTERNATIONAL SEARCH REPORT

ational Application No PCT. 98/01864

A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 C02F1/461 According to International Patent Classification (IPC) or to both national classification and IPC B. FIELDS SEARCHED Minimum documentation searched (classification system followed by classification symbols) C02F C25B Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched Electronic data base consulted during the international search (name of data base and, where practical, search terms used) C. DOCUMENTS CONSIDERED TO BE RELEVANT Citation of document, with indication, where appropriate, of the relevant passages Relevant to claim No. Χ US 4 784 735 A (SORENSON) 15 November 1988 Α see the whole document 1,5,7,9 EP 0 693 459 A (ALONE WORLD KK) Χ 24 January 1996 1.5 see column 7, line 57 - column 9, line 9 see column 10, line 2 - column 11, line 25 Α see figures 1.2 7.9 DE 22 27 084 A (HUELS CHEMISCHE WERKE AG) Α 13 December 1973 1,2,5-7,see figures 5,6: example 3 see page 4, paragraph 2 US 4 006 067 A (GUSSACK) 1 February 1977 1,2,5,7, see column 6, line 14 - line 57; figure Further documents are listed in the continuation of box \bar{C} Χ Patent family members are listed in annex. Special categories of cited documents "T" later document published after the international filing date "A" document defining the general state of the art which is not considered to be of particular relevance. or priority date and not in conflict with the application but cited to understand the principle or theory underlying the "E" earlier document but published on or after the international filing date "X" document of particular relevance; the claimed invention "L" document which may throw doubts on phority claim(s) or which is cited to establish the publication date of another cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone chation or other special reason (as specified) "Y" document of particular relevance; the claimed invention "O" document referning to an oral disclosure use, exhibition or cannot be considered to involve an inventive step when the document is combined with one or more other such docudocument published prior to the international filing date but ments, such combination being obvious to a person skilled later than the phority date claimed "&" document member of the same patent family Date of the actual completion of theinternational search Date of mailing of the international search report 29 September 1998 06/10/1998 Name and mailing address of the ISA European Patent Office, P.B. 5818 Patentlaan 2 Authorized officer NL - 2280 HV Rijswijk Tei (+31-70) 340-2040. Tx. 31 651 epo ni, Fax: (+31-70) 340-3016 Ruppert, G

Form PCT:ISA/210 (second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

· a	tional	Application No		
	ĢΒ	98/01864	ŀ	

	ation) DOCUMENTS CONSIDERED TO BE RELEVANT	
Category :	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No
A	US 4 964 970 A (O'HARE) 23 October 1990 see column 4, line 28 - line 57 see column 5, line 48 - line 51 see column 6, line 18 - line 37 see figure 2A	1,2,6,7,
4	US 3 984 303 A (PETERS ET AL) 5 October 1976 see the whole document	
Α	DE 195 17 652 A (GRUNDFOS AS) 21 November 1996 see figure 1	
İ		

1

Form PCT/ISA/210 (continuation of second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

	inf	ormation on patent family me	embers	ers PCT/	
Patent document cited in search repor	t	Publication date	Patent famil member(s)		Publication date
US 4784735	A	15-11-1988	NONE		
EP 0693459	. A	24-01-1996	JP 80248 AU 6928 AU 25082 US 56249	371 B 295 A	30-01-1996 18-06-1998 01-02-1996 29-04-1997
DE 2227084	Α	13-12-1973	NONE		·
US 4006067	Α	01-02-1977	NONE		. –
US 4964970	,A	23-10-1990	NONE		
US 3984303	A	05-10-1976	DD 1261 DE 26295 DK 2944	36 A 61 A 59 A 06 A 76 A 74 A,B, 53 A 26 A 98 A 01 A	26-07-1977 13-05-1980 30-04-1980 22-06-1977 20-01-1977 03-01-1977 28-01-1977 17-05-1978 21-01-1977 04-01-1977
DE 19517652	Α	21-11-1996	NONE		

THIS PAGE BLANK (USPTO)